

NEUTRON HOLOGRAPHIC STUDY OF PALLADIUM HYDRIDE

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Holography was invented by Dénes Gábor [1] in an attempt to construct an electron microscope attaining atomic resolution without image distortions due to electromagnetic lenses. Despite the widespread use of holographic techniques in the wavelength range of visible light, which followed the development of highly coherent laser beams, the original idea to record images on an atomic scale could be realized only about 40 years later. During the last decade holographic techniques based on electrons (i.e. matter waves) were successfully extended to electromagnetic waves (X-rays and γ -rays) readily available from synchrotron radiation. A major part of this work has been reviewed e.g. in [2].

Limitations in applying the above techniques arise in the case of electrons from their extremely strong interaction with condensed matter restricting them essentially to the investigation of surfaces. X-rays, on the other hand, while being able to penetrate more deeply into matter, exhibit variations of sensitivity covering several orders of magnitude over the periodic table impeding their use for many systems involving particular combinations of elements. Neutrons, in principle, are not subject to these drawbacks. However, for various reasons, partly related to the limited intensity of presently available neutron beams, their application was not considered feasible. Only recently [3] experimental setups were put forward permitting to transfer certain conceptions developed in the context of X-ray holography to the case of neutrons.

Holographic imaging techniques are based on the recording of the interference pattern of two coherent waves originating from the same source. The first wave, that reaches the detector directly, serves as the reference wave; the second one is scattered by the object of interest and subsequently interferes with the reference wave. In the context of holography with atomic resolution basically two techniques are in use which are called the *inside source* and the *inside detector* setup, respectively.

In the case of neutrons [3,4,5], the inside-source concept requires a point-like source of spherical neutron waves within the sample. Hydrogen nuclei (i.e. protons) are extremely well suited to serve this

purpose due to their large incoherent elastic scattering cross section (~ 80 barns) for thermal neutrons. Metal-hydrogen systems, therefore, are a natural choice in studying various features of this novel technique. The feasibility of *Neutron Inside Source Holography* was first demonstrated in an experiment on a mineral containing substantial amounts of hydrogen by a Canadian group at Chalk River [6] and is applied in the present experiment on the metal-hydrogen system PdH [7]. The second method, *Neutron Inside Detector Holography*, requires the application of strongly neutron-absorbing isotopes which act as point-like detectors within the sample. It was successfully applied by the present authors in an experiment at the ILL in Grenoble recording the holographic image of a lead single crystal containing a small fraction of neutron-absorbing cadmium [8].

In the present investigation a holographic image of palladium nuclei in a PdH_{0.78} single crystal was recorded [7]. The sample was a slightly irregular-shaped slab with the dimensions $\sim 2.8 \times 15 \times 8$ mm³. It was covered with a thin copper film whose thickness (~ 30 μ m) was sufficient to avoid hydrogen loss. Hydrogen is well known to occupy octahedral interstitial sites in the *fcc* palladium lattice. The palladium nuclei play the role of the object while the hydrogen nuclei (i.e. protons) serve as point-like neutron sources inside the sample.

The experiment was done with the goal to overcome certain technical problems in order to open the way towards a wider range of applications. A particular challenge of the experiment consisted in the extraordinarily strong variations of the scattering intensity due to the size and irregular shape of the sample exceeding the weak holographic modulation ($\sim 10^{-3}$) by several orders of magnitude. This problem, however, could be solved by the application of suitable filtering and image processing techniques. In addition, it was shown that the problem of Bragg peak contaminations encountered in the first inside-source measurements [6] can be completely avoided by choosing an experimental setup where the sample is rotated about two perpendicular axes

while the detector is kept fixed at a scattering angle where the momentum transfer does not fulfil the Bragg condition [5]. In the same way contributions of Debye-Scherrer rings due to the copper coating could be circumvented and, more generally, this technique can be applied to any sample environment.

The experiment was carried out at the 6T2 four-circle diffractometer at the LLB Saclay, using a neutron wavelength of ~ 0.9 Å. The sample was mounted on the cradle of the diffractometer and rotated about the angle χ through a range of 45° and about the angle ϕ through a range of 350° . The angular step-width was 5° for both rotations leading to a mesh of $10 \times 71 = 710$ pixels forming the hologram. Due to the strong scattering from hydrogen the mean free neutron path in the sample was only several *mm* and, consequently, the geometry of the sample is strongly influencing the recorded hologram (cf. Fig.1). However, since the wavelengths of the holographic intensity modulations are typically much shorter, the shape function can be removed by appropriate filtering without disturbing the holographic information. A mathematical reconstruction of the positions of the Pd atoms occupying crystal lattice sites around the hydrogen probe nuclei is shown in Fig.2.

The above results demonstrate that neutron holography indeed may serve as a novel tool to investigate structural properties of metal-hydrogen systems and can be expected to yield reliable information on the positions of atoms in crystal lattices. The experience gained in the present experiment suggests, however, that the availability of more sophisticated image processing techniques is likely to become an important issue in practical applications.

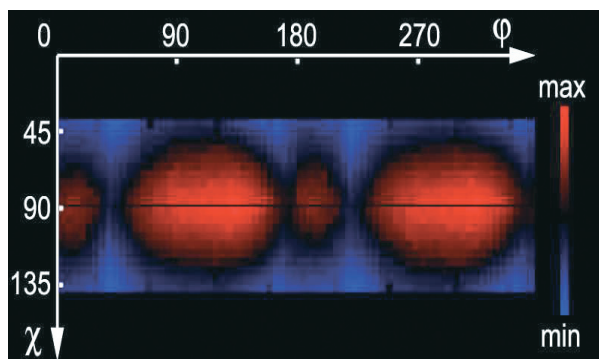


Figure 1. The raw hologram of the $\text{PdH}_{0.78}$ single crystal extended by symmetry operations to the whole range of the angle χ . The strong intensity variation due to the shape of the sample is clearly visible.

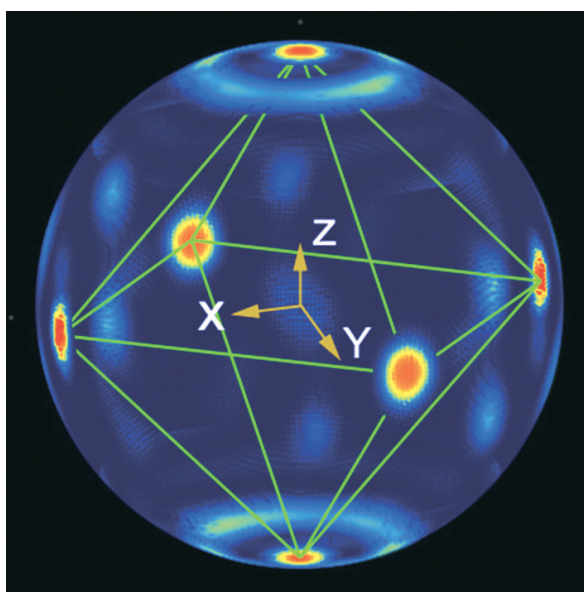


Figure 2. The restored holographic image of the local environment of a hydrogen nucleus. The three-dimensional octahedral arrangement of the Pd atoms is clearly visible. The hydrogen (not shown here) is located at the centre. The axes of the coordinate system are parallel to crystallographic [100] directions.

References

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